Decoherence and Absorption of $\text{Er}^{3+}:\text{KTiOPO}_4$ (KTP) at $1.5\ \mu\text{m}$

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We present results of laser absorption spectroscopy and two-pulse photon echo decoherence measurements on the lowest $^4\text{I}_{15/2}$ to lowest $^4\text{I}_{13/2}$ transition in $\text{Er}^{3+}:\text{KTiOPO}_4$ (KTP -potassium titanyl phosphate) for the optical transition located at 1537.238 nm. This transition was found to have an inhomogeneous absorption linewidth of 950 MHz and pronounced polarization dependence. Two-pulse photon echo decay measurements as a function of applied magnetic field strength at 1.9 K revealed a narrow homogeneous linewidth of 2.5 kHz at 0.2 T that increased to 5.8 kHz at 1.2 T and then decreased to 1.6 kHz at 4.5 T. This behavior was successfully described by decoherence due to $\text{Er}^{3+}-\text{Er}^{3+}$ magnetic dipole interactions. Significant superhyperfine coupling of Er^{3+} spins to the nuclear moments of ions in the host lattice was observed, modulating the photon echo decay at low magnetic fields and limiting the effective homogenous linewidth at high fields. Combined with the well-established potential of KTP for fabrication of high-quality optical waveguides and integrated non-linear frequency conversion, our results suggest that $\text{Er}^{3+}:\text{KTP}$ is a promising material system for practical spectral hole burning, signal processing, and quantum information applications.

Keywords: rare-earth-doped solids, decoherence, photon echoes, spectral diffusion, laser absorption spectroscopy, telecommunication wavelength

1. Introduction

The crystal KTiOPO_4 (KTP) is well-known for its unusually high nonlinear optical constants, moderate damage threshold, and wide transparency window. KTP is used extensively for frequency doubling of $1.06\text{ }\mu\text{m}$ Nd:YAG lasers [1,2], infrared optical parametric applications [3], electro-optic modulation, and waveguide devices [4]. Doping KTP with Er^{3+} ions has the potential to further enable many active optical devices that exploit the resonant optical transitions of Er^{3+} in the telecommunication window where commercial optical hardware is well developed. At cryogenic temperatures, Er^{3+} :KTP can also exhibit the phenomenon of spectral hole burning (SHB) that enables a wide range of information processing and storage applications in both the time and frequency domain. The unique potential of SHB materials has been demonstrated in GHz-scale time-domain signal processing, spatial-spectral holography, optical data storage, RF spectrum analysis, and laser frequency stabilization. SHB materials have also found applications in the field of quantum information science due to their low optical and spin decoherence rates, and we have recently suggested the potential of Er^{3+} :KTP as a promising system to support optically integrated quantum memories.[5] For Er^{3+} :KTP, transient spectral hole burning occurs for the lowest energy to lowest energy $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition of Er^{3+} that occurs near $1.54\text{ }\mu\text{m}$ by population storage in the $^4I_{13/2}$ excited state with a lifetime of more than 15 ms.[5] For example, using spectral holes in Er^{3+} :KTP as frequency references for diode laser frequency stabilization, we demonstrated an excellent short term fractional frequency stability of 1×10^{-12} over 1–100 ms averaging times and with long-term laser drift reduced to $\sim 200\text{ kHz}$ over 10 minutes. [6]

Among Er^{3+} doped materials operating at $1.5\text{ }\mu\text{m}$, Er^{3+} : Y_2SiO_5 and Er^{3+} : LiNbO_3 have so far shown the greatest promise, and have therefore been studied extensively and used in many proof of concept demonstrations. [5, 7-20] The Er^{3+} :KTP system, on the other hand, is still relatively unexplored. Initial photon echo measurements on Er^{3+} :KTP have revealed low decoherence rates [5], corresponding to narrow homogeneous linewidths in the frequency domain—properties that can make Er^{3+} :KTP suitable for applications in quantum information, SHB, and classical signal processing. Moreover, the combination of desirable optical coherence properties and the unique potential for integrated KTP waveguide devices interfaced with readily available off-the-shelf telecom hardware provide a strong motivation to investigate Er^{3+} :KTP further.

Only a few studies of Er^{3+} :KTP can be found in the literature [5, 21-23], leaving most of its spectroscopic properties, relaxation dynamics, and optical decoherence behavior unreported. In our initial study of Er^{3+} :KTP transitions near 1536 nm , we found that the spectrum contained absorption lines corresponding to at least 9 crystallographically inequivalent Er^{3+} sites.[5] In the work reported here, we focus on the strongest absorption line that occurs at 1537.238 nm and that offers the most promise for potential applications. We present new measurements on polarization dependent laser absorption and optical decoherence as a function of magnetic field strength. Photon echo decay measurements revealed the presence of decoherence due to spectral diffusion, a homogeneous linewidth broadening effect arising from spin-spin interactions between the Er^{3+} optical center and other electronic and nuclear spins in the crystal that cause time-dependent frequency shifts of the echo ions. Spectral diffusion can reduce the achievable

spectral resolution and information storage time in many applications, and hence must be characterized and understood to evaluate the capabilities and limitations of the material. We also present results that demonstrate significant superhyperfine coupling of Er^{3+} electronic states to the nuclear spins of the host.

2. Optical Absorption, Inhomogeneous Broadening, and Excited-state Lifetime

The Er^{3+} :KTP single crystal used in this work was prepared at the Clarendon Laboratory, University of Oxford and had a total Er^{3+} concentration of 0.04% atomic percent relative to Ti^{4+} , as determined from optical nutation, absorption, and emission measurements.[24] The crystal was transparent and appeared colorless with dimensions of $a = 12.87$ mm, $b = 5.18$ mm, and $c = 14.89$ mm. For laser absorption experiments, the crystal was held at a temperature of 3 K in a Cryomech PT405 cryocooler. The laser polarization at the sample was controlled using a zero-order half-wave plate and a Glan polarizer before the cryocooler. A homemade external cavity diode laser (ECDL) was used to provide a collimated beam with a ~ 1 mm waist and 14 μW optical power at the crystal for laser absorption measurements. The frequency of the laser was scanned by applying a linear voltage ramp to a piezoelectric transducer that drove the feedback prism plate of the laser. Laser light was coupled into a fiber Fabry-Perot with 49.4 MHz free spectral range to correct for small nonlinearities in the PZT scan and accurately calibrate the frequency sweep. The absolute vacuum laser wavelength was measured using a Burleigh WA-1500 wavemeter.

Laser absorption spectroscopy was carried out to investigate the inhomogeneous linewidth and polarization dependence of the lowest $^4I_{15/2} \rightarrow ^4I_{13/2}$ transitions. The spectrum of Er^{3+} :KTP near 1536 nm at cryogenic temperature is composed of a large number of sharp absorption lines that are associated with distinct Er^{3+} sites in the crystal. [5] While inhomogeneous broadening is caused by weak strain, significant charge and ionic-radius mismatch between the Er^{3+} dopants and the Ti^{4+} host ions can also introduce a range of larger perturbations that produce new “defect” lines in the Er^{3+} absorption spectrum. If we compare Er^{3+} :KTP to Er^{3+} :LiNbO₃, we find that the lattice imperfections in Er^{3+} :KTP primarily produce distinct defect lines, each with relatively narrow inhomogeneous linewidths, whereas in Er^{3+} :LiNbO₃ the lattice disorder results in a continuous distribution of broad absorption lines that overlap to produce an exceptionally large total inhomogeneous linewidth, although the scale and distribution of Er^{3+} energy perturbations appear to be nearly the same in both host crystals.

In this study, we investigated the properties of the main line at 1537.238 nm that offers the most promise for applications due to its larger absorption strength. From optical nutation measurements, it has been estimated that this site contains 49% of all Er^{3+} ions in the crystal, with the remaining 51% distributed among all of the other sites observed in the spectrum.[24] To determine the $^4I_{13/2}$ excited-state lifetime for this site, we directly excited the transition with a 40 ms pulse generated with a 80 MHz Brimrose fiber-coupled acousto-optic modulator (AOM) and detected the decay of the emitted fluorescence using a large-area amplified InGaAs photodiode with >1 MHz bandwidth. To minimize saturation of the excitation due to narrow-

band spectral hole burning, ~100 MHz of frequency noise was imposed on the excitation laser. The measured fluorescence decay is shown by the symbols in Fig. 1, revealing a long excited-state lifetime of $T_1 = 16.9$ ms. To verify that the measured lifetime was not affected by radiation trapping, we confirmed that the same decay time was observed at the peak of the absorption line and in the tails where the absorption is weak.

Figure 2 shows the polarized laser absorption spectrum for this site at $T = 3$ K for $\mathbf{E} \parallel \mathbf{c}$ and $\mathbf{E} \parallel \mathbf{a}$ with the laser propagating along the $\mathbf{b}$ -axis ($\mathbf{k} \parallel \mathbf{b}$). The laser absorption spectrum reveals an inhomogeneous linewidth of 950 MHz (FWHM) with a peak absorption of 3.5 cm^{-1} for $\mathbf{E} \parallel \mathbf{c}$ and 1.1 cm^{-1} for $\mathbf{E} \parallel \mathbf{a}$. From additional measurements with $\mathbf{k} \parallel \mathbf{a}$, we also found a peak absorption of 1.7 cm^{-1} for $\mathbf{E} \parallel \mathbf{b}$. While these three measurements would be sufficient to characterize the absorption for electric dipole transitions, the magnetic dipole transition matrix elements are often comparable to the electric dipole matrix elements for the $^4I_{15/2} \rightarrow ^4I_{13/2}$ transitions of Er^{3+} . As a result, six independent measurements with different combinations of $\mathbf{E}$ and $\mathbf{k}$ vector directions (corresponding to all orthogonal combinations of optical $\mathbf{E}$ and $\mathbf{B}$ vectors) are generally required to fully determine the transition probabilities for arbitrary conditions. Nevertheless, we may make a rough estimate for the transition oscillator strength using our measured laser absorption spectra and assuming electric dipole transitions, indicating a value of $f \sim 1 \times 10^{-7}$. If we combine the oscillator strength estimate for this transition with the relative emission strengths to all of the multiple $^4I_{15/2}$ levels determined from the fluorescence spectrum for this site, we calculate an excited-state radiative lifetime of ~22 ms, an estimate that is remarkably close to the measured value of 16.9 ms from Fig. 1.

Our measurements also revealed a significant broadening and distortion of the Er^{3+} absorption spectrum in some regions near the surfaces of the KTP crystal. An extreme example of this effect is presented in Fig. 3 for absorption lines near the 1537.238 nm transition where the linewidths broaden and distinct side peaks appear for some of the absorption lines. Considering that a relatively large spot size of ~1 mm was used in these measurements and that the broadening and distortion is not observed when the beam passes through regions further within the crystal, the side peaks that appear in the spectrum appear to originate from overlap of the beam with small regions near the surface that exhibit distinct shifts of the transition energies. This unusual effect indicates a significant perturbation of the crystal lattice near the surface, potentially arising from deviations from ideal stoichiometry due to out-diffusion of K^+ or mixed crystal phases at the surface, both of which are known effects for KTP. [25,26] In particular, we expect that the formation of inhomogeneities near the crystal surface depends primarily on the details of the crystal cutting, polishing, and any thermal annealing or chemical treatment processes used when preparing a sample from the crystal boule. Since waveguide applications of KTP exclusively probe the region of the crystal near the surface, it is essential to better understand these anomalous effects. Consequently, additional studies on new crystals will be required to determine whether this spatial inhomogeneity is unique to our particular sample or something that occurs more widely in rare-earth-doped KTP

3. Optical Decoherence and Spectral Diffusion

The homogeneous linewidth in the frequency domain and the corresponding phase coherence lifetime in the time domain are crucial material parameters relevant for SSH, SHB, and quantum information applications. Achieving useful homogeneous linewidths and coherence lifetimes requires the application of a magnetic field to suppress spectral diffusion due to Er^{3+} spin flips by depopulating the ground-state's upper Zeeman sub-level. Often placing the sample between permanent rare-earth magnets is sufficient to accomplish this task. [6]

To determine the rate of phase decoherence and the associated homogeneous linewidth, parameters that are typically hidden to conventional methods because of limited spectral resolution, we performed two-pulse photon echo measurements on the main absorption line at 1537.238 nm as a function of magnetic field strength. For these measurements, the crystal was held at $T = 1.9$ K in a superfluid liquid helium bath using an Oxford Instruments Spectromag cryostat capable of generating magnetic fields of up to $B = 8$ T. The liquid helium temperature was accurately determined from the measured vapor pressure. The laser's $\mathbf{k}$ -vector was parallel to the crystal's $\mathbf{b}$ -axis while the magnetic field and the laser's $\mathbf{E}$ -field were both parallel to the crystal's $\mathbf{a}$ -axis. The ECDL output power of ~ 1.8 mW was amplified by an erbium-doped fiber amplifier. A 165 MHz AOM gated the amplified laser beam to generate pulses for two-pulse echo excitation with dynamic range of >30 dB. Pulse peak powers at the sample were ~ 3 mW with a focused beam waist of $20 \mu\text{m}$ and the pulse widths were ~ 500 ns for the first pulse and ~ 700 ns for the second pulse. The >1 MHz pulse bandwidths minimized sensitivity of the photon echo signal to laser frequency fluctuations at the longest pulse delays. A gating AOM after the crystal was used to discriminate the echo signal from the excitation pulses. The echo signal was detected using a fast amplified InGaAs photodiode and processed with a digital oscilloscope interfaced with a data-acquisition computer that recorded the integrated area of the echo signal as a function of the pulse separation in the two-pulse excitation sequence.

Photon echo decays were measured for magnetic field strengths varied from $B = 0.15$ T to $B = 5$ T. Figure 4 shows representative echo decay curves for several magnetic field strengths. The observed echo decays were non-exponential, indicating time-dependent decoherence due to the presence of spectral diffusion during the echo pulse sequence. We analyzed the echo intensity using the empirical Mims equation [27]

$$I(t_{12}) = I_0 \cdot \exp \left\{ 2 \left(\frac{2t_{12}}{T_M} \right)^x \right\}, \quad (1)$$

where I_0 is the initial echo intensity, t_{12} is the separation between pulse 1 and pulse 2, and x is an empirical parameter that depends on the spectral diffusion dynamics that determine the shape of the echo decay curve. The phase coherence lifetime T_M obtained using Eq. 1 is the time over which the initial intensity decays to I_0/e^2 . Solid lines in Fig. 4 are fits to Eq. 1 and show excellent agreement with the data. The effective homogeneous linewidth Γ_h over the timescale of the echo decay measurement can be defined in terms of T_M using the relation

$$\Gamma_h = \frac{1}{\pi T_M}. \quad (2)$$

In the absence of spectral diffusion, echo decays are exponential with a Mims exponent of $x=1$ and $T_M = T_2$ so that the effective homogeneous linewidth defined in Eq. 2 becomes equal to the single-ion homogeneous linewidth.

Figure 5 shows the variation of the effective homogeneous linewidth Γ_h as a function of magnetic field strength. Narrow linewidths of several kilohertz were observed for all field strengths. However, the variation of the effective homogeneous linewidth with magnetic field strength shows a substantial increase at intermediate fields that appears as a “bump” in the overall trend. At a very low magnetic field of $B = 0.2$ T we measure $\Gamma_h \sim 2.5$ kHz, increasing with magnetic field to a maximum value of ~ 5.8 kHz at $B = 1.2$ T, and then decreasing to a minimum value of 1.6 kHz at $B = 4.5$ T. Further increases in magnetic field strength do not produce any additional decrease in the effective homogeneous linewidth.

The magnetic field dependence can be quantitatively modeled using established theories for the magnetic dipole-dipole interaction between Er^{3+} ions.[8, 28] Because Er^{3+} ions exhibit large electronic magnetic moments, they are particularly sensitive to local magnetic field fluctuations in the crystal. For example, whenever an Er^{3+} ion in the crystal changes its electronic spin, all nearby Er^{3+} ions in its local environment are affected. Time-dependent perturbations of each ion’s transition frequency due to the dynamic nature of the ion’s environment gives rise to spectral diffusion that manifests itself in non-exponential photon echo decays that reflect an evolution of the decoherence rate over time. Following our original detailed analysis of spectral diffusion in $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$, [8] we can apply the same models to describe the observed behavior in $\text{Er}^{3+}:\text{KTP}$. In this model, we describe the magnetic field and temperature dependent spectral diffusion linewidth Γ_{SD} using

$$\Gamma_{\text{SD}}(B, T) = \Gamma_{\text{max}} \text{sech}^2 \left(\frac{g_{\text{env}} \mu_B B}{2kT} \right), \quad (3)$$

where Γ_{max} is the maximum FWHM frequency broadening due to magnetic dipole-dipole interactions, g_{env} is the g value of the perturbing magnetic moments, μ_B is the Bohr magneton, B is the magnetic field strength, k is Boltzmann’s constant, and T is the temperature. As can be seen from the above equation, the spectral diffusion linewidth is suppressed for larger magnetic fields and lower temperature as the spins preferentially align with the external magnetic field.

The Er^{3+} spin flips can be driven by a variety of processes, such as direct phonon absorption or emission, elastic and inelastic phonon scattering, and mutual Er^{3+} – Er^{3+} spin flip-flops, with each mechanism exhibiting a different characteristic magnetic field and temperature dependence. Following our treatment for $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$, we find that the spectral diffusion rate in $\text{Er}^{3+}:\text{KTP}$ can be best described by the form

$$R(B) = R_0 + \alpha_D g_{\text{env}}^3 B^5 \coth \left(\frac{g_{\text{env}} \mu_B B}{2kT} \right), \quad (4)$$

where R is the characteristic rate of the spectral diffusion process given by the sum of the upward and downward spin-flip transition rates of the perturbing spins. The first term R_0 represents the magnetic field independent contributions, such as Raman and Orbach phonon scattering processes. The second term represents the one-phonon direct process for spin-lattice relaxation involving absorption or emission of a single phonon resonant with the perturbing moment's spin-flip transition, where α_D is a generally anisotropic constant characterizing the strength of the phonon coupling. The rate R rapidly increases with magnetic field because the available phonon density of states increases with energy, while on the other hand Γ_{SD} rapidly decreases with an increase in magnetic field strength due to an increase in magnetic order as the thermal population in the upper Zeeman component of the ground state gets frozen out. It is the opposing effects of magnetic field strength on R and Γ_{SD} that are responsible for the observed local maximum, or “bump”, in the field dependence of the effective homogeneous linewidth Γ_h . [10] We have observed a similar magnetic field dependence of the effective homogeneous linewidth in a number of Er^{3+} doped materials, including Y_2SiO_5 [9, 10] and 2G2S glass. [29] Accordingly, this spectral diffusion “bump” appears to be quite common in a broad range of Er^{3+} doped materials, as well as other systems with large magnetic moments in general where spectral diffusion due to magnetic dipole-dipole interactions is present.

To quantitatively model the magnetic field dependence, we may examine the influence of R and Γ_{SD} on the phase decoherence time T_M . Following Refs. [8, 28], the phase decoherence time T_M for two-pulse echoes can be calculated from the material parameters Γ_0 , Γ_{SD} , and R as

$$T_M = \frac{2\Gamma_0}{\Gamma_{SD}R} \cdot \left(-1 + \sqrt{1 + \frac{\Gamma_{SD}R}{\pi \Gamma_0^2}} \right), \quad (5)$$

where Γ_0 is the homogeneous linewidth in the absence of spectral diffusion and all other parameters have the same meaning as before. Fitting Eq. 2 with T_M given by Eq. 5 to the data gives the solid line in Fig. 5, showing excellent agreement with the measured values and quantitatively explains the observed behavior. In our fit, we used the measured values for the ground state g value of $g_{\text{env}} = 12$ for $\mathbf{B} \parallel \mathbf{a}$ and $T = 1.9$ K as fixed parameters while the four unknown parameters Γ_0 , Γ_{max} , R_0 , and α_D were allowed to vary to produce the fit to the data in Fig. 5 with values of $\Gamma_0 = 1.6$ kHz, $\Gamma_{\text{max}} = 8.8$ MHz, $R_0 = 3$ Hz, and $\alpha_D = 3.3 \times 10^{-4}$ kHz/T⁵.

The magnetic field dependence in Fig. 5 reveals a surprisingly weak spectral diffusion broadening at very low magnetic field strengths of only ~ 2.5 kHz. For $B < 0.2$ T, the direct phonon process becomes slow enough that the behavior in Fig. 5 is primarily governed by the R_0 term in Eq. 5 at those low fields. Because the weakest field studied in this work was $B = 0.15$ T, the precise value of R_0 is not very well determined and could in fact be significantly slower than the value of 3 Hz extracted from the fit. If we compare the spectral diffusion broadening at low fields to other materials such as $\text{Er}^{3+}:\text{LiNbO}_3$ or $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$, we find that the value of R_0 in $\text{Er}^{3+}:\text{KTP}$ is at least two orders of magnitude slower. Furthermore, there is insufficient data to definitively distinguish between a field independent R_0 that results from two-phonon scattering, a weak field-dependent term due to Er^{3+} - Er^{3+} mutual spin flip flops, or a combination of both

mechanisms.[8] Even if we account for differences in Er^{3+} concentration and dipole-dipole interaction strengths, the Er^{3+} - Er^{3+} spin flip-flop rate would be expected to be much faster than what is observed, indicating that the flip-flop process that usually dominates Er^{3+} spectral diffusion behavior at very low fields is unusually weak in this material. This suggests that Er^{3+} :KTP may be an ideal candidate for persistent hole burning by optical pumping of electronic spin states (Zeeman sublevel storage) [30] since the value of R_0 effectively establishes the maximum lifetime for that mechanism.[8]

For low magnetic fields of $B < 0.2$ T, we observed a strong field-dependent modulation in the two-pulse echo decays. An example of this behavior is shown in Fig. 6 for $B = 0.15$ T at $T = 1.9$ K. This echo modulation is a well-known effect that has been observed in many different material systems [7, 9, 31 - 37] and can be attributed to superhyperfine coupling between Er^{3+} electronic states and neighboring nuclear spins in the host lattice. The echo modulation at these low magnetic fields occurs since the echo pulse sequence is able to simultaneously excite transitions between multiple superhyperfine levels that can coherently interfere, producing a complex modulation in the echo intensity as the excitation pulse spacing is varied. The modulation may be analyzed using the approach of Mims [32] to extract information about the superhyperfine level splittings and identify which nuclear spins are coupled to the Er^{3+} electronic states. The solid line in Fig. 6 is a fit of the echo modulation model to the data indicating that the data can be approximately described using superhyperfine splittings of 88 kHz and 175 kHz for the two electronic levels involved in the transition. The corresponding effective gyromagnetic ratios of 0.587 MHz/T and 1.17 MHz/T suggest that the observed superhyperfine interaction is likely due to coupling of Er^{3+} to neighboring ^{39}K nuclear spins in the lattice. The large chemical shifts of the observed effective nuclear gyromagnetic ratios relative to the free nucleus value of 1.9868 MHz/T for ^{39}K [38] indicate a large Er^{3+} - ^{39}K interaction strength. Additional systematic measurements as a function of magnetic field strength and orientation are required to confirm this hypothesis. In particular, the study of spin-flip sidebands in hole burning spectra can provide information regarding interactions with the nuclear spins.

Further evidence for superhyperfine coupling between Er^{3+} and host ion nuclear spins was given by the behavior of the observed echo decay curves shown in Fig. 4 for the high magnetic field strengths. The shapes of the echo decays for $B = 4.5$ T and $B = 5$ T are identical and field independent, yielding the same phase decoherence time while still remaining non-exponential. This general behavior has been observed for transitions in many other materials that contain large nuclear spins, such as $\text{Er}^{3+}:\text{YLiF}_4$, $\text{Er}^{3+}:\text{LaF}_3$, and ruby. [39 - 44] At these high field strengths, Er^{3+} - Er^{3+} magnetic dipole-dipole interactions can be completely suppressed so that the remaining optical decoherence is caused by superhyperfine interactions with fluctuating nuclear spins in the host and therefore approaches a constant value referred to as the superhyperfine limit. As a result, the superhyperfine interactions experienced in KTP affect spectral diffusion dynamics, produce coherence modulation, and ultimately limit the minimum homogeneous linewidths and decoherence rates that may be achieved.

4. Conclusion

We studied the main optical absorption line in bulk Er^{3+} :KTP at 1537.238 nm and found an excited-state lifetime of 16.9 ms, an inhomogeneous linewidth of 950 MHz, and a magnetic field dependent effective homogeneous linewidth of a few kilohertz. The observed decoherence was successfully modeled using spectral diffusion theory for direct phonon-driven magnetic dipole-dipole interactions between the Er^{3+} ions. Furthermore, low and high magnetic field data revealed superhyperfine coupling of the Er^{3+} ions to the nuclear magnetic moments of the host ions.

While this study focused on the dominant Er^{3+} site at 1537.238 nm that is most relevant for potential device applications, a complete understanding of the complex defect structure of Er^{3+} :KTP will require comprehensive site-selective spectroscopy measurements across all of the observed absorption lines. In addition, our results suggest the need for further study of how the surface structure and chemistry of KTP is affected by the sample fabrication process to better understand and minimize variations in the absorption spectrum near the surface of the crystal. This is particularly important for waveguide applications since different approaches for waveguide fabrication and annealing may have important effects on the observed spectroscopic properties.

Together, our findings are important and promising results for the application of Er^{3+} :KTP in quantum memory and signal processing applications at 1.5 micron telecom wavelengths.

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Figure captions

Figure 1. Measured unpolarized fluorescence decay (symbols) of 0.04% Er^{3+} :KTP for direct excitation of the most strongly absorbing Er^{3+} site at 1537.238 nm for $T = 3$ K. Fitting the exponential decay (solid line) reveals a $^4I_{13/2}$ excited-state lifetime of 16.9 ms for this site.

Figure 2. Polarized laser absorption spectrum of 0.04% Er^{3+} :KTP for the most strongly absorbing Er^{3+} site at 1537.238 nm at $T = 3$ K. The laser was propagated with $\mathbf{k} \parallel \mathbf{b}$ and the laser's electric field was oriented $\mathbf{E} \parallel \mathbf{c}$ (solid line) or $\mathbf{E} \parallel \mathbf{a}$ (dashed line). The absorption line was fitted to give an inhomogeneous linewidth of 950 MHz with peak absorption of 3.5 cm^{-1} and 1.1 cm^{-1} for $\mathbf{E} \parallel \mathbf{c}$ and $\mathbf{E} \parallel \mathbf{a}$, respectively.

Figure 3. Laser absorption spectra at 1537.238 nm for two different positions in the 0.04% Er^{3+} :KTP crystal at $T = 3$ K with $\mathbf{k} \parallel \mathbf{b}$ and $\mathbf{E} \parallel \mathbf{c}$. Throughout most of the crystal volume, the spectrum shown by the solid line is observed; however, in some regions near the surface of the crystal the spectra become noticeably broadened and distorted, as shown by example spectrum represented by the dotted line, perhaps indicating excess strain, mixed crystal phases, or deviations from ideal stoichiometry near the surface.

Figure 4. Two pulse photon echo decays in 0.04% Er^{3+} :KTP at $T = 1.9$ K as a function of magnetic field strengths with $\mathbf{B} \parallel \mathbf{a}$, $\mathbf{E} \parallel \mathbf{a}$, and $\mathbf{k} \parallel \mathbf{b}$.

Figure 5. Effective homogeneous linewidth dependence on magnetic field strength in 0.04% Er^{3+} :KTP at 1.9 K with $\mathbf{B} \parallel \mathbf{a}$, $\mathbf{E} \parallel \mathbf{a}$, and $\mathbf{k} \parallel \mathbf{b}$. Symbols correspond to experimental data and the solid line is a fit using the spectral diffusion model for Er^{3+} - Er^{3+} magnetic dipole-dipole interactions.

Figure 6. Two pulse photon echo decay in 0.04% Er^{3+} :KTP at $B = 0.15$ T at $T = 1.9$ K with $\mathbf{B} \parallel \mathbf{a}$, $\mathbf{E} \parallel \mathbf{a}$, and $\mathbf{k} \parallel \mathbf{b}$. The strong observed modulation can be attributed to the coupling of Er^{3+} to neighboring nuclear spins in the host lattice. The solid line is a fit to the data including the effects of superhyperfine coupling.

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